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Methodology for Estimating the Production and Release of Airborne Radionuclides Produced in Accelerator and Beamline Enclosures

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Abstract

A method for estimating airborne releases of radionuclides, due to operation of an accelerator facility, in both enclosed recirculated air volumes and vented air volumes is described. The methodology is an extension of that presented in Fermilab-TM-2089, motivated by the need to calculate releases due to leakage from highly activated enclosed volumes of air into larger volumes of vented air. This methodology has been reviewed and approved by the Fermilab ES&H Section for application to the design, construction and operation of the NuMI facility.

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1 Introduction

The operation of most accelerators and beamlines will result in the production of airborne radionuclides due to the interaction of the primary and secondary beam particles with air. The result of experience at high energy proton accelerators is that the short-lived radionuclides ^{11}C, ^{13}N, ^{41}Ar and ^{15}O as well as ^{3}H are produced. Other radionuclides are also produced, but are not a concern for environmental or personnel protection.

Due to higher beam intensities, levels of air activation near target regions become quite high. As a result, it is prudent to enclose and recirculate this highly activated air, allowing only small amounts to leak out to the larger vented volume of air that is then released to the environment.

In this report we extend the methodology presented in FERMILAB-TM-2089, "Production and Release of Airborne Radionuclides Due to the Operation of NuMI" to the case where an enclosed volume leaks to an outer volume that is then vented to the environment.

2 Summary of Regulatory Requirements

Federal regulations, which are further implemented by the State of Illinois, govern the releases of airborne radionuclides, excluding radon and radon progeny, by U. S. Department of Energy facilities (CFR89). These regulations place an annual limit of 10 mrem in a year on the dose equivalent that can be delivered to a member of the public due to the release of airborne radionuclides from DOE facilities. The methodology for determining the dose equivalent is also specified. The regulations further require the application of continuous monitoring in accordance with U. S. Environmental Protection Agency specifications if the dose equivalent should exceed 0.1 mrem in a year. Maintaining the total site boundary dose equivalent to be less than 0.1 mrem in a year permits the Laboratory to measure these releases as a part of a confirmatory monitoring program. It has been standard practice for the Fermi National Accelerator Laboratory to be considered as a single facility under this regulation. Thus, these limits apply to the total release from the Laboratory.

3 Determining Site Boundary Dose Equivalent Rates

A dose equivalent rate of 0.1 mrem/year or less, approximately distributed uniformly throughout several months, cannot feasibly be measured directly. The alternative indirect method used to estimate such a dose equivalent consists of measurements of the activity released from the various sources using stack monitors at the release points. The activity released, expressed in terms of the individual radionuclides found at the stacks, is then used as input in the computer code CAP88-PC along with specified meteorological data for the calendar year during which the releases occurred[1]. The computer code, required by the applicable regulations (CFR89), calculates the dose equivalent delivered at the Fermilab site boundary. The results fluctuate from year-to-year due to details of the Fermilab operational schedule and variations in meteorological parameters used as input data. During CY 1996, 21 Ci were released from the Laboratory and a dose equivalent at the site boundary of 0.013 mrem was calculated. During this period, the Fermilab Fixed Target program was operating with 800 GeV protons along with simultaneous

[1] The Fermilab ES&H Section performs these calculations.

operations of the Antiproton source that used 120 GeV protons. Similar operations during CY 1997 resulted in 0.015 mrem dose equivalent due to the release of 29.5 Ci. The average of these results for the site boundary yields a dose equivalent per unit activity of 5.64×10^{-4} mrem/Ci, a scaling factor that can be used to roughly predict dose rates at the site boundary.

4 Fermilab Air Pollution Operating Permit

Consistent with these requirements, in March 1999, Fermilab submitted an application renewal to the Illinois Environmental Protection Agency for its lifetime air pollution operating permit[2]. This application addressed the radionuclide emissions from NuMI, and other Fermilab facilities. It specified that the doses to the public will be kept well below 0.1 mrem/year for all Fermilab operations. The average annual activity release is to be kept less than 100 Ci[3].

5 Modeling of the Beamline

The primary beam transport and secondary beam transport is done using the MARS simulation program (Mo98). The user input to the program includes parameters describing the proton beam, layout of the beamline elements and their geometry and the surrounding shielding and enclosure geometry. The user can also define to what energy level particles are tracked, depending on the specific application. The output from MARS simulations that is needed for this air activation methodology is the charged and neutral hadronic flux densities. The cross sections for production of radionuclides in air are relatively independent of energy above a few ten's of MeV. Thus for MARS flux density estimations to be used in this methodology, a threshold of 20 MeV is recommended. The MARS flux densities determined in the various air volumes in the enclosure are then used, with the appropriate cross sections, to determine the air activation levels for various isotopes.

6 Methodology Including a Leakage Term

The production of airborne radioactivity has been discussed elsewhere in detail (Co99, Ref. TM-2089) and thus will not be covered here. The isotopes of concern are the short-lived radionuclides ^{11}C, ^{13}N, and ^{15}O as well as ^{3}H. ^{7}Be is also produced, but is thought to plate out on various beamline elements, as it has never been detected in the releases from target stations at Fermilab. It is therefore not further considered with respect to the release of airborne radioactivity to the environment. ^{41}Ar has a large cross section for production by thermal neutrons. It is very difficult and time consuming to estimate the thermal neutron flux densities using MARS, and thus a scaling factor is used. The value of the concentration of ^{41}Ar is conservatively taken to be 2.5 % of the sum of the concentrations of ^{11}C and ^{13}N calculated within a given enclosure. This choice is conservative in that typical measured values at Fermilab are 1%, although 2.5% has been measured. ^{41}Ar, ^{13}N and ^{11}C tend to be the radionuclides of concern, although this methodology also calculates the levels of ^{15}O and ^{3}H. The short half-life of ^{15}O and the small decay constant of ^{3}H usually result in insignificant levels of these two radionuclides.

[2] In response to this application, Illinois Environmental Protection Agency permit was issued on June 16, 1999.

[3] The ES&H Section has the details of the air permit, where such things as maximum allowable releases are specified.

In some cases one may need to model a separate volume that is enclosed (hereafter called the confined volume), but leaks at some rate into a second volume. The second volume is the volume that is then vented or released to the environment. The same general methodology is used as described previously, but with a slight variation.

One assumes that the activity in the vented volume is in equilibrium. The same calculation is done in the confined volume, where now the ventilation rate is the leakage rate and the transit time is zero, so there is no decay in transit term. The activity is then multiplied by the leakage rate over the ventilation rate (of the outer volume) and added in to the first term in equation 1 (below). In this way the two steps of ventilation (leakage and then the release to the stack) are taken into account accurately.

Thus, using the air release rate, confined volume leakage rate, flux densities and volumes (see Table 1), one can calculate the total release, in terms of activity, of each individual radionuclide over some period of time. One does this by multiplying this specific activity, by the volume of air released during the same period (D_y); in this case a year. These are also summed over the various radionuclides, i, to get the total activity (Ci/year) released per year at the stack, a_y (equation 1 below). In some applications the outer vented air volume, which is not highly activated, is comparable to the volume of air to the vent. Since the highly activated air being released from the confined volume, once released to the outer vented volume can then decay, it may be reasonable to use a fraction, F, of the vented volume in calculating the transit time (and thus decay) to the vent. The value of F depends on the particular application. If there is no confined volume and the outer volume is the source of activated air, then F should be set to 0.

Equation 1:

$$a_y = DO_s N_p \sum_{i,j}\left[\left(N_j\sigma_i\right)\left(\phi_{reg} + \phi_{conf}\frac{D_l}{D}\left(\frac{\lambda_i}{\lambda_i + r_l}\right)\left(1 - e^{-(\lambda_i + r_l)t_{irrad}}\right)\right)\right]\left(\frac{\lambda_i}{\lambda_i + r}\right)\left(1 - e^{-(\lambda_i + r)t_{irrad}}\right)\left(e^{-\lambda_i(t_{transit} + t_{cool})}\right)$$

where,

N_j is the number of target atoms per unit volume (atoms/cm^3).

σ_i is the cross section for production of radionuclide *i* from target atom *j* (mb).

ϕ_{reg} is the average flux density in the vented region as determined from MARS (particles/cm^2/proton).

ϕ_{conf} is the average flux density in the confined region, determined from MARS (particles/cm^2/proton).

N_p is the number of incident protons per second at the source, averaged over the year.

λ_i, is the inverse mean lifetime of radionuclide *i* in units consistent with those of r, and the irradiation and cooldown times.

t_{irrad} is the irradiation time of the air volume

r the ventilation term, is the number of air changes per unit time.

$$r = \frac{D}{V_T}, \text{ and}$$

$$V_T = \left(V_{reg}F + V_{vent}\right)$$

D is the ventilation rate in the vented air volume per unit time.
F is the fraction of V_{reg} one considers when calculating the ventilation term r.
V_{reg} is the volume of air in the vented volume.
V_{vent} is the volume of the air from V_{reg} to the release point.
r_l the leakage term, is the leakage rate per unit time. The leakage volume is the confined air volume that is leaking to the outer vented volume.

$$r = D_l / V_{conf}$$

V_{conf} is the volume of air in the confined volume.
D_l is the confined volume leakage rate in consistent units of volume per unit time.
$t_{transit}$ is the ventilation system travel time, from the production region to the release point:

$$t_{transit} = \frac{V_T}{D} = \frac{1}{r}$$

t_{cool} is the time after beam-off at which one is calculating the release; typically this is zero as one calculates for a continuous beam-on release.
O_s is the number of operational (beam-up) seconds year.

N_j, σ_i, and λ_i, are given in Section 10 and do not change for different applications. Table 1 shows the parameters specific to the accelerator or beamline that are needed to estimate the radioactive air emissions.

Table 1: Input Parameters for Air Activation Calculations

Parameter	Units	Comments
ϕ_{reg}, ϕ_{conf}	Hadrons/cm^2/p	From MARS simulations
N_p	Protons/sec	Typically average over 3 years
D, D_l	cm^3/sec	Outer released air volume release rate, and inner captured volume leakage rate (respectively)
V_{reg}, V_{vent}, V_{conf}	cm^3	Outer released air volume, air volume from released volume to stack, and inner confined volume (respectively)
t_{irrad}	sec	Irradiation time of air volume
t_{cool}	sec	Cooldown time of air volume
F	unitless	Fraction of activated volume of air included in determining the transit time of the air to the release point
O_s	seconds	Operational (beam-up) time per year, typically 60% up time is assumed

7 Dose Equivalent Rate to Personnel within the enclosure due to Exposure to Activated Air

This methodology can be used to estimate the dose equivalent rate due to exposure to activated air within the beamline enclosure. As observed in TM-2089, this dose equivalent rate to personnel accessing an area with radioactive air rapidly decays to trivial levels after only about an hour of cooldown. This is due to the short half-lives of most of the radionuclides produced.

An estimate of the radiological hazard to a person present in this air can be obtained by comparison with the derived air concentration (DAC) for ^{3}H. Other radionuclides are ignored

since they have such short half-lives. Working in air containing radionuclides at one DAC corresponds to an effective dose equivalent rate of 2.5 mrem hr^{-1} (including internal deposition of ^{3}H). The DAC value for ^{3}H in the form of elemental hydrogen is 5 x 10^{-7} Ci cm^{-3} (CFR93). It is also straightforward to calculate the dose equivalent rate that might be received by a worker accessing the "enclosure volume" after a given cooldown period.

If one wishes to estimate the dose equivalent to a worker entering the enclosure after a shorter period than 1 hour, then all the radionuclides should be considered. In this case the concentrations found, after uniformly mixing the contributions of the regions, are used to calculate the effective dose equivalent rate due to exposure to this air. These concentrations correspond to the first two terms in brackets in equation 2 below. This is done by taking the concentrations for each radionuclide of concern, including ^{7}Be, calculated at the end of the irradiation, $C_i(0)$, dividing them by the appropriate Derived Air Concentrations (DACs) for occupational workers, and summing them to determine the total fraction of a DAC achieved (see discussion of DACs in TM-2089).

Equation 2:

$$a_{i,reg} / cm^3 = \sum_j \left[N_j \sigma_i \phi_{reg} \left[\left(\frac{\lambda_i}{\lambda_i + r} \right) \left(1 - e^{-(\lambda_i + r) t_{irrad}} \right) \left(e^{-\lambda_i t_{cool}} \right) \right] \right]$$

In general, the effective dose equivalent rate, $dH(t_{cool})/dt$, as a function of decay time, t_{cool}, is thus given by,

$$\frac{dH(t_{cool})}{dt} = 2.5 \sum_i \frac{C_i(0)}{DAC_i} \exp[-(\lambda_i + r) t_{cool}] \text{, mrem hr}^{-1}),$$

since the exchange of air reduces the concentration of activity in a manner similar to that provided by the radioactive decay.

8 Discussion of Uncertainty

It is useful to have an idea as to the uncertainty on the values of Ci/year released out a stack in order to understand how high the value might be above what this method estimates. The uncertainty in the final result depends on the geometry of the particular situation. If a shield pile has all of its iron covered with concrete or some other material that will absorb neutrons, then the 2.5% factor for ^{41}Ar is most likely over-estimated and one should look at what the result would be with a more typical scaling factor of 1%. Uncertainties on the cross sections for the production of the radionuclides of concern are 25%. Uncertainties in the hadronic flux densities from MARS, assuming accurate geometry modeling, are approximately 25%. Since the thermal neutron production of ^{41}Ar is added add hoc, detailed modeling of cracks in the shielding and

detailed chemical analysis of the shielding material are not necessary in MARS. It is reasonable to determine the Ci/year release rate out the stack for several realistic scenarios of either ^{41}Ar content, ventilation rates and/or leakage rates from the confined volume. This then provides a range of possible results that could be obtained when operating.

9 Summary

A methodology for estimating airborne radionuclide release rates from a stack has been presented. The methodology incorporates leakage from a sealed internal volume to an outer volume that then releases the air out a stack. ^{41}Ar production is added in adhoc due to the complexities of determining an accurate thermal neutron flux density. Some rough guidelines are given for extrapolating this value to a dose rate at the site boundary. In practice, CAP88 must be used to accurately predict the value. The radionuclides of concern have short half-lives and thus if one waits an hour before accessing the beamline or accelerator enclosure, the dose to personnel due to airborne activation will most likely be minimal.

10 Physical Parameters for Typical Radionuclides Produced in Air at a High Energy Proton Accelerator

Table 2 gives values of decay constants and production cross sections. The production cross sections are conservative choices taken from the references by Barbier (Ba69) and Thomas and Stevenson (Th88).

Table 2 Physical Parameters for Typical Radionuclides Produced in Air at a High Energy Proton Accelerator

	Decay Constants, λ_i, and High Energy Cross Sections, σ_{ijHE} (mb) for the Production of Various Radionuclides					
Product =>	**^{3}H**	**^{7}Be**	**^{11}C**	**^{13}N**	**^{15}O**	**^{41}Ar[4]**
λ (sec^{-1}) =>	1.79×10^{-9}	1.51×10^{-7}	5.69×10^{-4}	1.16×10^{-3}	5.67×10^{-3}	1.05×10^{-4}
Target Nuclide	**High Energy Hadron Cross Sections, σ_{HE} (mb)**					
^{14}N	30	14	20	4	0	
^{16}O	30	8	10	5	35	
^{40}Ar	1	10	1	1	1	
^{15}N	30	14	20	4	0	
^{18}O	30	8	10	5	35	

[4] The production of ^{41}Ar by means of thermal neutron capture is handled in an *ad hoc* manner. See discussion in the text.

11 Acknowledgements

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